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Release of C-14 and H-3 from Irradiated Graphite of the Thermal Column of VVR-S Reactor to Gas Phase

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^{14}C and ^{3}H are produced in all types of reactors, mainly through neutron-induced reactions with common isotopes of carbon, nitrogen and oxygen. They are present in the fuel, fuel cladding, coolant, moderator and structural materials of the reactor. Irradiated graphite contains wide range of activation products but, from the point of view of geological disposal, two radionuclides have to be taken into account: tritium (^{3}H or T) and carbon-14 (^{14}C). The release and migration of carbon ^{14}C and ^{3}H are critical issue for geological disposal of high/intermediate level radioactive waste. In this paper we present some results obtained regarding the release of ^{14}C and ^{3}H, for a relatively long time period into gas phase from intact samples of irradiated graphite which have been cut from the thermal column of the VVR-S reactor from Horia Hulubei National Institute for Physics and Nuclear Engineering (IFIN-HH). An experimental apparatus has been designed and built to measure the release of ^{14}C and ^{3}H to gas phase from irradiated graphite. The experimental results regarding the release of ^{14}C and ^{3}H to gas phase show that only a small fraction of the initial inventory was released in a 12-month period. The result obtained should be applied with caution for a long time prediction.

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PACS/topics: irradiated graphite, release, tritium, carbon-14

1. Introduction

The VVR-S nuclear reactor from IFIN-HH is a research reactor with a maximum thermal power of 2 MW using distilled light water as moderator, coolant and reflector. The reactor was commissioned in 1957 and was operated with nuclear fuel type EK-10 (enrichment 10%) and from 1984, this fuel was replaced by a new fuel type S-36 (enrichment 36%) that was used until 1997, when the reactor was definitively shut-down and now is under decommissioning. Figure 1 shows a horizontal cross section of the VVR-S reactor core [1].

Generally, the decommissioning of reactors will involve management and disposal of a big amount of operational waste. Except for the reactor core, the main activation products in the reactor come from the graphite and building materials. Even though, the nuclear grade graphite is a relatively pure material and the concentration of the most part of impurities is at ppm level there are still enough impurities able to produce numerous radionuclides by activation. ^{14}C is produced in the reactor graphite due to the presence of precursor nuclide ^{13}C, ^{14}N (main contributor) and ^{17}O.

The main contribution to accumulation of ^{3}H in the reactor graphite results from the neutron activation reaction $^{6}\mathrm{Li}(\mathrm{n},\alpha)^{3}\mathrm{H}$.

In the case of VVR-S reactor the overwhelming amount of radionuclides contained in the graphite thermal column is represented by ^{14}C and ^{3}H. Therefore the potential release of ^{14}C and ^{3}H from graphite requires particular attention for long term storage.

^{14}C and ^{3}H are both beta emitters (half-life 5730 years long lived radionuclide and half-life 12.3 years relatively short-lived radionuclide, respectively). As a consequence, the release ^{3}H is important only for intermediate storage safety of the disposal facility, which is not the case for the release of ^{14}C containing gases from irradiated graphite [2–4].

2. Experimental procedure

The mobile thermal column (Fig. 1) is made of 6 graphite discs and each disc is cladded in aluminum. Initially, the thermal column was provided with a cooling system that axially penetrated the graphite plate connected to the water-cooling system. During the reactor operation, it was concluded that the cooling system was no longer necessary and it was given up. The horizontal tubes were filled with graphite rods of the same type as the graphite discs of the thermal column.

The irradiated graphite samples were taken from these nuclear grade graphite rods.

TABLE I

Main characteristic of the irradiated graphite samples used for gas release experiment.

Graphite sample	No. of discs	Mass [g]	C-14 inventory [Bq]	H-3 inventory [Bq]
6.1.2	2	2.548		
6.2.1	2	1.982	179861.5	593671.4
6.2.2	2	2.109		

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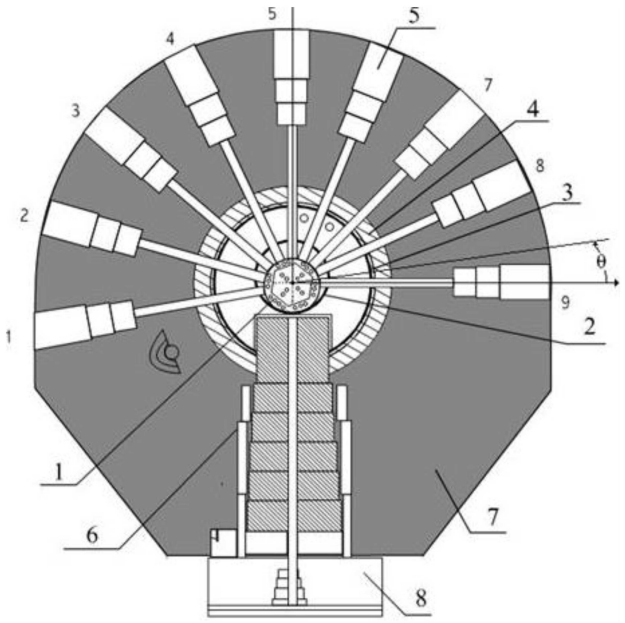

Fig. 1. Vertical cut-view through the VVR — S reactor and the movable thermal column, 1 — reactor core, 2 — median vessel, 3 — outer vessel, 4 — cast-iron rings, 5 — beam tubes, 6 — thermal column, 7 — biological shield, 8 — thermal column shielding.

For this experiment the graphite sampling was performed from a small graphite rod located around the central graphite rod of the disc no. 6 located near the reactor core. One piece was cut from the rod edge near the reactor vessel (Sample 6.1) and the others two pieces from the opposite part of the rod (Sample 6.2.1 and 6.2.2) (see Table I).

The ^{14}C and ^{3}H specific activities of the irradiate graphite used in the experiment were determined by a full combustion method followed by isotopes separation and liquid scintillation counting. The results have been presented in a previous paper [5].

The irradiated graphite samples were sealed in a glass vessel provided with 4 outlets and inlets. Forty days after the samples were sealed; the vessel was connected to a catalytic oxidizer furnace that contains a CuO catalyzer inside. The ^{14}C and ^{3}H compounds released in the gas phase were oxidized in a low oxygen flow rate atmosphere and trapped separately in 5 vials (Fig. 2). At the end of the measurement the vessel containing the graphite samples was resealed and the experiment was repeated after 2 and 12 months. The first two trapping vessels each contained 5 ml of sulfuric acid 0.1 N (for tritium trapping) and the following three vessels each contained 5 ml of Carbo-Sorb E (for carbon-14 trapping). The sulfuric acid solutions in the first two vessels were mixed together. An aliquot (2 ml of retention solution) was transferred to a 20 ml LSC vial, and 16 ml of ULTIMA GOLD cocktail was added. The same procedure was followed for the vessels containing Carbo-Sorb, but this time Permafluor scintillator was used. ^{14}C and ^{3}H activity was measured by a TRICARB TR 2800 liquid scintillation counter. The sample and the blank were measured for 100 min and 3 cycles, and the averages of three cyclic measurements were calculated [6].

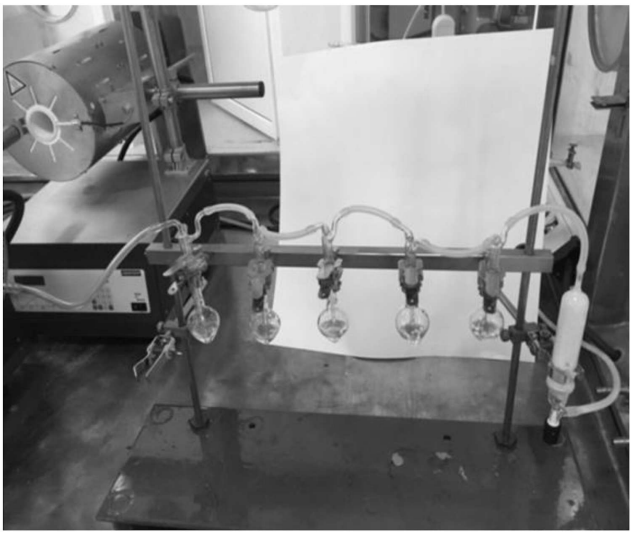

Fig. 2. Apparatus for C-14 and H-3 determination release in gas phase.

3. Results and discussion

Based on the results of carbon-14 and tritium activities and the dilution factors, the total carbon-14 and tritium activities released into gas phase were calculated.

The total carbon-14 and tritium activity released into gas phase are presented in Table II.

TABLE II

C-14 and H-3 activities released into gas phase.

Sampling period [days]	C-14 activity		H-3 activity	
	total	background	total	background
	[Bq]		[Bq]	
14	0.9±12%*	0.5±0.3**	0.8±12%*	0.3±0.2**
60	12.2±12%*	0.5±0.3**	10.9±11%*	0.3±0.2**
365	116.5±10%*	0.5±0.3**	59.5±11%*	0.3±0.2**

* — overall uncertainty — estimated.

** — standard counting deviation.

4. Conclusion

The experiments to measure the release of carbon-14 and tritium into gas phase were performed over a 12-month period. The irradiated graphite was collected from the thermal column of the VVR-S Reactor (at IFIN-HH). Graphite samples were taken from the graphite rod located near the reactor core (graphite disc no. 6). The total mass of the samples was 6.639 g. The initial inventory of carbon-14 and tritium is shown in Table I.

An experimental apparatus has been designed and manufactured to measure the total release of carbon-14 and tritium to gas phase from irradiated graphite samples collected from the thermal column of the VVR-S Reactor.

The total carbon-14 and tritium released to gas phase after 365 days represent approximately 0.064% and respectively 0.010% of the estimated inventory in the graphite samples.

However, these results should be applied with caution for a long-time prediction.

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